Apatite from granitic pegmatite exocontacts in Moldanubian serpentinites

Apatit v exokontaktech granitických pegmatitů v moldanubických serpentinitech (Czech summary)

(2 figs)

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Geochemical features of apatite from three contact assemblages of granitic pegmatites intersecting serpentinized peridotites were examined to test their dependence on the character of the pegmatites, which are considerably modified by reaction with the ultrabasic host rocks. The cation content of all three apatite samples does not deviate from the usual Ca-rich composition with only negligible Fe, Mn, Mg, Na and traces of Si, U, Th and Pb. However, the REE and Y contents are quite variable among the three occurrences. In the oligoclase+phlogopite assemblage of the footwall contact of the Věžná I pegmatite, the $Fap_{81}OHap_{18}Clap_1$ apatite contains minor Y, REE, Fe and Mn, conforming to the presence of Y- and REE-bearing minerals in the parent beryl-columbite-subtype pegmatite body. In the phlogopite rimming simple plagioclasite veinlets at Věchnov, the REE-poor, HREE- and Y-depleted $OHap_{59}Clap_{28}Fap_{13}$ with Fe>Mn and slightly elevated Sr reflects the geochemically primitive signature of the pegmatites, mobilized from adjacent gneissic lithologies. At Heřmanov, the HREE-depleted $Fap_{81}OHap_{17}Clap_2$ associated with phlogopite and minor actinolite is slightly Sr- and Ba-enriched but otherwise rather close to the composition of the Věžná I apatite. This and other evidence suggests that the adjacent quartzo-feldspathic rock may represent a primitive rare-element pegmatite rather than a migmatitic mobilizate.

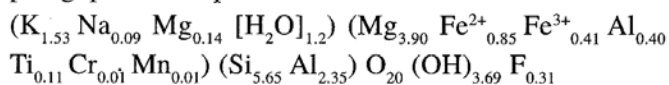*Key words:* apatite, granitic pegmatite, serpentinite, rare-earth elements, Moldanubicum

Introduction

Despite the extensive attention paid to apatite from all petrological environments (e. g., Povondra 1992), there are virtually no data available for apatite from contact-reaction assemblages of granitic pegmatites in ultrabasic country rocks, peridotites and serpentinites. The note by Babu (1972) seems to be a unique exception. Considering the great extent to which granitic pegmatites are locally modified in such host rocks, becoming desilicated and alkali-depleted, chemical composition of apatite deserves examination of its response to the exotic combination of these ultrabasic and silicic environments, and of the overall signature of apatite which may betray the character of the pristine, pre-reaction pegmatite melt. We report here on apatite from contact assemblages of three pegmatites crosscutting serpentinite bodies in the Strážek Moldanubicum of western Moravia: Věžná I, Věchnov and Heřmanov (Fig. 1).

Věžná I

The Věžná I pegmatite crosscuts an orthopyroxene peridotite, metamorphosed to an amphibole- and chlorite-bearing facies and subsequently extensively serpentinized, south of the village of Věžná, west of Nedvědice. The pegmatite is classified as the beryl-columbite subtype of the rare-element class, with niobian rutile, beryl, zircon, xenotime-(Y), monazite-(Ce) and brabantite as the most common accesory minerals, and abundant beryllian cordierite as the main internal product of contamination (Černý – Novák 1992). Most of the blocky feldspar has

K/Rb values between 210 and 146, but the K-feldspar adjacent to extremely rare pods with, *i. a.*, lepidolite and pollucite shows K/Rb as low as 88 to 52 (Novák – Pelz 1981, Teertstra et al. 1995). A layered sequence of anthophyllite, actinolite and phlogopite separates the serpentinite wallrock from the pegmatite. Along the central parts of the footwall contact of the pegmatite, a phlogopite + oligoclase (An_{18}) unit is located (Černý – Miškovský 1966) with abundant apatite dispersed in the phlogopite matrix and in part embedded in the subsurface of the coarse crystals of oligoclase. Composition of the phlogopite corresponds to:
$(K_{1.53} Na_{0.09} Mg_{0.14} [H_2O]_{1.2}) (Mg_{3.90} Fe^{2+}_{0.85} Fe^{3+}_{0.41} Al_{0.40} Ti_{0.11} Cr_{0.01} Mn_{0.01}) (Si_{5.65} Al_{2.35}) O_{20} (OH)_{3.69} F_{0.31}$
(Černý – Miškovský 1966); composition of the oligoclase averages at ~3.5 wt. % CaO, equivalent to An_{18} (Černý et al. 1984).

On optical-microscopic and electron-microprobe (EMPA) examination, pale apple-green apatite is homogeneous and unzoned, but it contains sporadic inclusions of other phases. The inclusions are largely 3 to ~ 30 μm in size, and in many cases cannot be reliably identified because of probable extension of the analyzed volume from the tiny inclusions into the apatite matrix. Xenotime and zircon were identified, and a number of inclusions of what seems to be a REE-rich apatite with highly variable composition.

Věchnov

Small, less than 10 cm wide veins of plagioclasite crosscut an enstatite- and amphibole-bearing serpentinite close
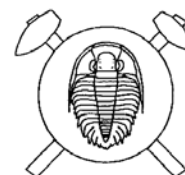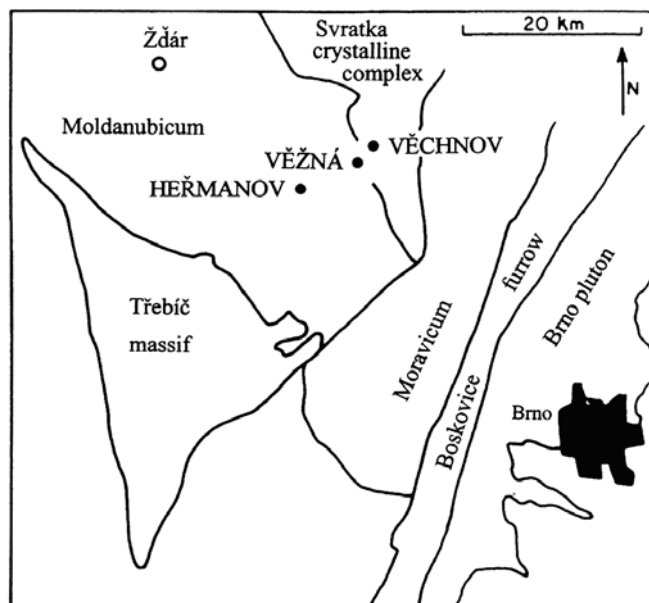
Fig. 1 Location map of the three occurrences of granitic pegmatites examined in the present study.

to the village of Věchnov, east of Rožná at Bystřice nad Pernštejnem. The veins are virtually monomineralic, consisting of oligoclase-andesine An_{23-28} which is largely decomposed into chlorite, saponite and other undefined clay minerals. Quartz is absent; very rare and in part altered grains of perthitic K-feldspar have K/Rb of 260 to 235. A thick layer of considerably weathered phlogopite adjoins the plagioclasite, with lesser amounts of actinolite and anthophyllite toward the ultrabasic wallrock. Several prismatic crystals of apatite were found embedded in the phlogopite.

Pale apple-green apatite is homogeneous on optical-microscopic and EMPA scales, and free of inclusions of other phases.

Heřmanov

The apatite described here comes from the well-known locality of the "Anthophyllit-Glimmerkugeln", rounded nodules of radial anthophyllite with phlogopite cores (André 1804, Brezina 1874), embedded in phlogopite matrix.

The phlogopite forms schlieren with more or less pronounced preferred flake orientation in a pegmatitic rock, which consists of perthitic K-feldspar (K/Rb 83 to 72), dominant over oligoclase (An_{13-17}) and subordinate quartz, minor diopside, accessory zircon and probable molybdenite (M. Novák, pers. comm. 1999). Serpentinite is not exposed at the locality or in its close neighbourhood, and even Suess (1895), in the most detailed account of petrographic relationships available in the literature, characterizes this assemblage as a pegmatite in gneiss. However, the asemblage of anthophyllite + actinolite + phlogopite and the distinctive Cr and Ni contents of the phlogopite leave no doubt about the contact-metasomatic origin of these phases along a pegmatite interface with an ultrabasic rock. The latter may be hidden at depth, or it could have been completely digested by the silicic melt (Černý 1972).

Apatite and minor, greenish-coloured actinolite are disseminated in phlogopite, which has the composition of $(K_{1.73} Na_{0.08} Rb_{0.01} Ca_{0.008} Ba_{0.001}) (Mg_{1.15} Fe^{2+}_{0.31} Ti_{0.17} Fe^{3+}_{0.07} Cr_{0.05} Li_{0.04} Ni_{0.02}) (Al_{1.89} Fe^{3+}_{0.14} Si_{5.97}) O_{19.87} (OH)_{3.36} F_{0.77}$ (Černý 1972). The pale-greenish apatite is homogeneous on optical-microscopic and EMPA scales, but it contains extremely rare microinclusions of a REE-rich phase, possibly apatite (≤ 10 μm in size).

Experimental methods

Electron-microprobe analysis (EMPA) of apatite was performed on the Cameca SX–50 instrument at the Department of Geological Sciences, University of Manitoba, in the wavelenght-dispersion mode with beam diameter of 1 to 2 μm, acceleration potential of 15 kV, sample current of 20 nA and counting time of 20 s (40 s for F, Cl, Ba, Sr, U, Th and Y). The standards and analytical lines used here were the same as in Smeds et al. (1998). Data were reduced using the PAP procedure of Pouchou – Pichoir (1984, 1985).

The REE, Y, Sr, Ba, U, Th and Pb were analyzed by the inducively coupled plasma – mass spectrometer (ICP-MS) instrument at the Metals Laboratory, Institute for Environmental Reserch, University of Windsor. Powdered apatite samples (nominally 0.1 g) were dissolved in screw-top Teflon PFA bombs in 8 N HNO_3, evaporated to dryness and redissolved in 0.1 N HNO_3. The apatite samples, as well as replicates of the SRM SY-4 (dissolved according to the procedure of Jenner et al. 1990) were analyzed at dilutions of approximately 1:40 000. Element sensitivities and interelement correction factors were determined using the two standard solutions (STDA and STDB) of Jenner et al. (1990), diluted 100fold. Internal drift was corrected by use of the internal standards Be, In and Tl. Precision of the REE, based on replicate analysis of SY-4, was less than 5 %, and accuracy less than 8 %.

Unit-cell dimensions were refined in hexagonal setting from X-ray powder diffractograms calibrated with annealed synthetic CaF_2 ($a = 5.45935$ Å), recorded on a Philips PW 1710 apparatus with $CuK\alpha_1$ radiation. The FIX program of Ercit (1986), which compensates for the $\alpha_1 - \alpha_2$ interference, and the CELREF program of Appleman – Evans (1973) were used for the unit-cell refinements.

Refractive indices were measured in immersion liquids using the Becke line method and a monochromatic light source ($\lambda = 589$ nm).

Chemical composition

Table 1 summarizes the chemical composition of the apatite samples examined. In general, all three apatite compositions are mutually similar in terms of cation contents, with only minor variations at individual localities. Fe and Mn are generally low, as is Mg, Na and Pb. Silicon is negligible, and Al even more so; U and Th are at the detec-

Table 1 Chemical composition of the examined apatite samples as determined by electron-microprobe analysis; atomic contents normalized to 13 anions, H_2O calculated by stoichiometry.

	VZ	VC	H
P_2O_5	41.90	41.67	41.79
SiO_2	0.02	0.01	0.12
UO_2	0.01	0.02	0.03
ThO_2	0.00	0.02	0.04
Al_2O_3	0.00	0.01	0.01
As_2O_3	0.00	0.00	0.00
Bi_2O_3	0.02	0.01	0.03
Y_2O_3	0.27	0.01	0.11
FeO	0.10	0.10	0.08
MnO	0.10	0.04	0.08
CaO	54.37	54.07	53.26
SrO	0.04	0.30	0.94
MgO	0.02	0.12	0.07
PbO	0.03	0.03	0.02
Na_2O	0.09	0.02	0.21
F	3.02	0.48	3.02
Cl	0.05	1.94	0.14
H_2O	0.32	1.04	0.29
O=F	−1.27	−0.20	−1.27
O=Cl	−0.01	−0.44	−0.03
Total	99.08	99.25	98.94
atoms per formula unit			
P	3.006	3.006	3.011
Si	0.002	0.001	0.010
Σ IV	3.008	3.007	3.021
U	0.000	0.000	0.001
Th	0.000	0.000	0.001
Al	0.000	0.001	0.001
As	0.000	0.000	0.000
Bi	0.000	0.000	0.001
Y	0.012	0.000	0.005
Fe^{2+}	0.007	0.007	0.006
Mn	0.007	0.003	0.006
Ca	4.936	4.937	4.856
Sr	0.002	0.015	0.046
Mg	0.003	0.015	0.009
Pb	0.001	0.001	0.000
Na	0.015	0.003	0.035
Σ	4.983	4.982	4.967
F	0.809	0.129	0.813
Cl	0.007	0.280	0.020
OH	0.183	0.590	0.167
O	12.000	12.000	12.000

Table 2 REE contents and selected trace elements of the examined apatite samples as determined by ICP-MS.

	VZ	VC	H
La	350	432	1002
Ce	212	769	873
Pr	161	96	417
Nd	707	372	1996
Sm	251	67	584
Eu	9	11	80
Gd	271	45	336
Tb	50	4	35
Dy	313	21	158
Ho	58	4	25
Er	151	10	61
Tm	19	1	8
Yb	105	7	44
Lu	13	1	6
Y	2110	103	737
Sr	393	1952	7567
Ba	9	17	225
Pb	62	31	142
Th	9	6	49
U	39	49	94

subhorizontal distribution in the Věžná I apatite close to 1000x chondritic, with La/Yb of 3.3 (La/Lu 28) but prominent negative anomalies for Ce and Eu. The two anomalies are rather similar but weaker for the Heřmanov sample, which shows slight decline in HREE abundances, progressive with increasing atomic number (La/Yb 23, La/Lu 175). However, the pattern is distinctly different for the Věchnov apatite: the Eu anomaly is negligible and that of Ce virtually non-existent, and the slope toward low HREE and Y contents very distinct (La/Yb 61, La/Lu 420).

Physical properties

Unit-cell dimensions and refractive indices of the apatite samples examined are quoted in Table 3. All data are in good agreement with the values expected for the given chemical compositions according to the correlations published by Winchell – Winchell (1951), McConnell (1973), Tacker – Stormer (1989), Hughes et al. (1989, 1990) and Nesse (1991).

Discussion

Crystal chemistry

The apatite samples examined do not show any prominent deviations from the ideal tetrahedal population of P, and the Ca site also is closely conformable to the ideal formula. The substitution for Ca is very limited, and restricted mainly to divalent cations. The contents of Na, Si, U, Th and Al are, however, too low for any attempt at deciphering substitution mechanisms responsible for their incorporation.

The Věžná I and Heřmanov samples with dominant F, subordinate (OH) and mere traces of Cl should possess the normal hexagonal P6_3/m structure, but the sample from Věchnov is a candidate for an (OH)-Cl ordered

tion limit of the electron microprobe. The only exceptions are the Y contents of the Věžná I and Heřmanov apatite, and the elevated Sr content of the Heřmanov sample.

The anionic content is somewhat more diversified. The Věžná I and Heřmanov samples both correspond to (OH)-enriched fluorapatite with negligible Cl, whereas the Věchnov mineral is chlorian hydroxyapatite with subordinate F.

REE contents

Table 2 shows the contents of REE's, Y, Sr, Ba, Pb, U, and Th as determined by ICP-MS on bulk separates of the apatite samples. The Sr, U and Th data are in good agreement with those determined by EMPA (Table 1). The chondrite-normalized abundances of REE's and Y (Fig. 2) show
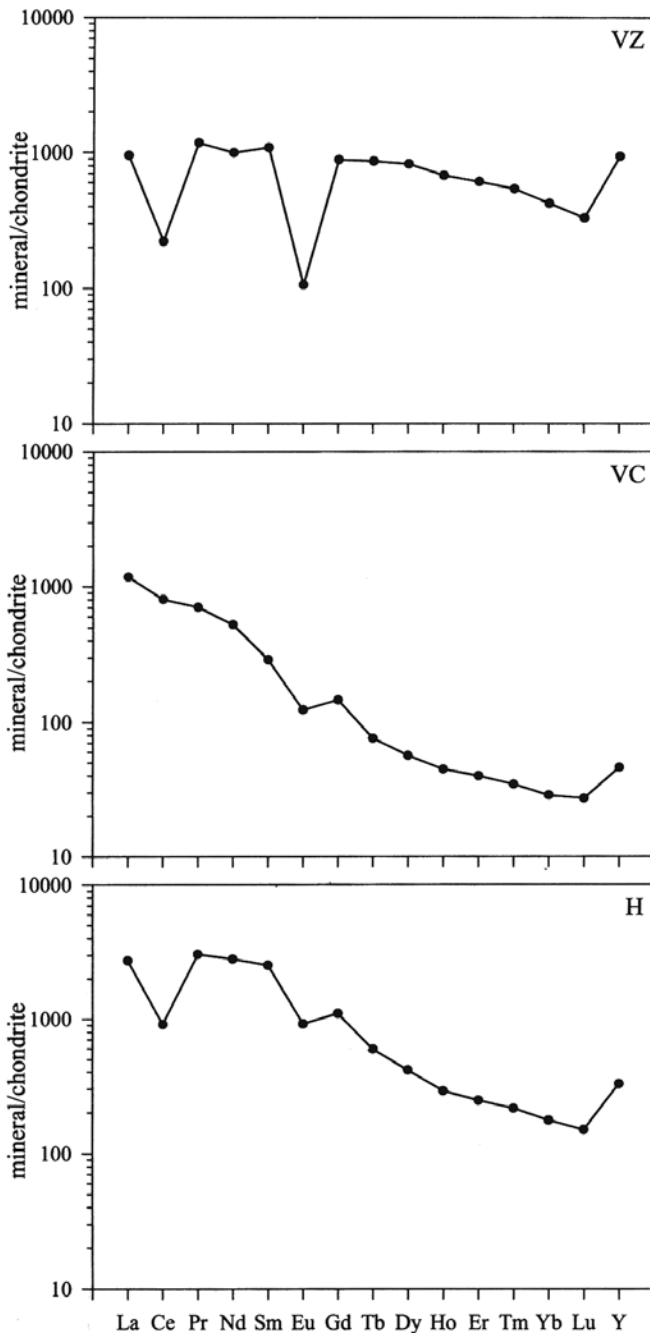
Fig. 2 Chondrite-normalized REE + Y data for the apatite samples examined: Věžná I (VZ), Věchnov (VC), and Heřmanov (H). Chondritic values from Taylor – McLennan (1985).

Table 3 Optical properties and unit-cell dimensions of the examined apatite samples.

	VZ	VC	H
ε	1.633	1.649	1.633
ω	1.637	1.654	1.638
ε–ω	0.004	0.005	0.005
a(Å)	9.385(1)	9.481(2)	9.388(2)
c(Å)	6.834(1)	6.838(2)	6.835(1)

is compositionally variable among different crystals, and that the chemical composition and physical properties established in 1967 must have been determined on fragments of different compositions. The 1967 anionic composition is so much different from the present data that an error in labelling the 1967 laboratory samples must also be considered.

Geochemical signatures and genetic significance

Compared to paragenetic sequences of apatite from highly evolved granitic pegmatites (such as those documented by Jolliff et al. 1989), the REE + Y contents and abundance patterns of the three apatite samples examined here are rather simple. Nevertheless, the data show some unusual features and help to interpret the nature of the pegmatite host rocks.

The enhanced REE content of the Věžná I apatite and its elevated HREE + Y contents are in agreement with the presence of accessory zircon and xenotime in the parent pegmatite. Actually, some of the HREE and Y contents of this apatite, as analyzed by ICP-MS, probably come from microinclusions of these minerals commented on in the introductory description. However, the EMPA data indicate that a substantial proportion of Y is contained in the apatite itself.

The negative Ce anomaly well expressed in the chondrite-normalized patterns of the Věžná I and Heřmanov samples is encountered in the literature only exceptionally. In the pegmatitic rocks of the Bohemian massif, Povondra (1992) found this anomaly only in apatite from the Přibyslavice granite. Puchelt – Emmermann (1976) reported the negative Ce anomaly from several unrelated rock types, and offered simultaneous crystallization of monazite (or other Ce-dominant phases) as an explanation. Monazite-(Ce) and brabantite do occur in the Věžná I pegmatite, but are paragenetically separated from the apatite examined; no Ce-bearing phases are known from the Heřmanov locality. Thus the above interpretation does not seem to be valid, but we have no alternate suggestion.

The general similarity of the Věžná I and Heřmanov chondrite-normalized patterns suggests that the Heřmanov quartzo-feldspathic rock, adjacent to the apatite-bearing phlogopite + actinolite assemblage, may represent a partially desilicated, geochemically primitive rare-element pegmatite rather than a migmatitic neosome mobilized from regional gneissic lithologies. Such a conclusion is also

structure with the monoclinic space group $P2_1/b$ (Hughes et al. 1990). However, a refinement of the crystal structure would be required to verify this possibility.

A special note must be devoted to the Věchnov apatite. It was described by Černý (1967) as a fluorapatite with subordinate (OH) component and traces of Cl, but with refractive indices and unit-cell dimensions which did not match such chemical composition. Our present data on chemistry and physical properties, collected on material from a different crystal, are internally consistent. The physical properties obtained in 1967 deviate from those expected for a F-dominant apatite toward the present data. All this indicates that the Věchnov apatite

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supported by the relatively low K/Rb value of the K-feld-spar. Rare-element pegmatites hosted by serpentinites in the west-moravian Moldanubicum generally postdate ser-pentinization of their host rocks (Černý – Povondra 1965); lack of alteration of the primary minerals of the Heřmanov assemblage corresponds with such timing.

In contrast, the mineral assemblage and extensive Mg-metasomatism of the plagioclase core of the Věch-nov pegmatite correspond to an early pegmatite genera-tion described in detail from other localities (Černý – Povondra 1965). Mineralogy of these pegmatites is very simple, and their geochemical signature very primitive; they were injected into the ultrabasic rocks prior to the conclusion of serpentinization, and are best correlated with partial melting and migmatite formation in the sur-rounding gneissic terrane. Chemical composition of the Věchnov apatite conforms to this interpretation, which is also suggested by the high K/Rb ratio of the K-feldspar. The REE content in general, and particularly that of HREE and Y is low, with negligible negative anomalies, and the prominent Cl content suggests partial melting in the presence of metamorphic pore fluids rather than an evolved, differentiated medium corresponding to a rare-element pegmatite melt. Chlorine-enriched apatite is otherwise typical of late, secondary stages of pegmatite evolution or of some low-temperature hydrothermal as-semblages (e. g., Fransolet et al. 1985, Elton 1997).

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Apatit v exokontaktech granitických pegmatitů v moldanubických serpentinitech

Byla studována geochemická charakteristika apatitu ze tří kontaktních asociací granitických pegmatitů protínajících serpentinity, za účelem ověření jejich závislosti na kategorii pegmatitu, které jsou značně modifikovány reakcí s ultrabazickou boční horninou. Obsah kationtů všech tří apatitů se neodchyluje od obvyklého složení s převahou Ca; obsahy Fe, Mn, Mg, Na jsou velmi nízké, a Si, U, Th a Pb jsou pouze ve stopách. Obsahy REE a Y jsou však na jednotlivých lokalitách dosti rozdílné. Apatit v asociaci s oligoklasem a flogopitem podél podložního kontaktu pegmatitu Věžná I odpovídá složení $Fap_{81}OHap_{18}Clap_1$ s minoritním Y, REE, Fe, a Mn; složení obráží přítomnost minerálu Y a REE v mateřském beryl-columbitovém pegmatitu. Flogopit lemující jednoduché plagioklasitové žilky u Věchnova obsahuje apatit o složení $OHap_{59}Clap_{28}Fap_{13}$ s Fe>Mn a zvýšeným Sr, chudý na REE a ochuzený yttriem a lehkými vzácnými zeminami. Toto složení odpovídá geochemicky primitivní signatuře mateřského pegmatitového žilníku, mobilizovaného z okolních rulových litologií. U Heřmanova, na lokalitě známých „heřmanovských koulí“, se apatit vyskytuje společně s aktinolitem ve flogopitu přilehlém k pegmatitovému tělesu. Apatit má složení $Fap_{81}OHap_{17}Clap_2$ s poněkud zvýšenými obsahy Sr a Ba a nízkou koncentrací HREE, jinak se však blíží apatitu z Věžné I. Tato okolnost a další ukazatelé naznačují, že přilehlá pegmatitová hornina pravděpodobně reprezentuje primitivní pegmatit vzácných prvků, spíše než migmatitový mobilizát.